Topology-Induced Spin Frustrations at the Cr(001) Surface Studied by Spin-Polarized Scanning Tunneling Spectroscopy

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The magnetic structure of the Cr(001) surface was investigated by spin-polarized scanning tunneling spectroscopy by making use of the spin-polarized surface state located close to the Fermi level. Periodic alternations of the intensity of the surface state peak in local tunneling spectra measured above different ferromagnetic terraces separated by monatomic steps confirm the topological antiferromagnetic order of the Cr(001) surface. Screw dislocations cause topology-induced spin frustration, leading to the formation of domain walls with a width of about 120 nm.

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The magnetic structure of the Cr(001) surface has been of great interest already for a long time because of the existence of surface ferromagnetic order despite the antiferromagnetic bulk structure [1,2] and because of the important role of the magnetic properties of Cr(001) in exchange-coupled Fe/Cr bilayers and superlattices [3,4]. In 1989 Blügel *et al.* [2] recognized the influence of surface defects, such as monatomic steps, on the overall magnetic properties of the Cr(001) surface. The presence of such monatomic steps leads to a topological antiferromagnetic order with neighbored terraces exhibiting a magnetization in opposite directions. This microscopic model of the Cr(001) surface was confirmed one year later by spin-polarized scanning tunneling microscopy (SP-STM) [5,6]. In this experiment, CrO_2 thin film tips with a high degree of spin polarization were successfully used to detect periodic alternations of the measured monatomic step heights in constant-current images. The deviations of the measured step height values from the topographic monatomic step height could directly be related with the effective spin polarization of the tunneling junction. A significant drawback of this experimental approach was the superposition of topographic and magnetic structure information. To solve this problem, SP-STM has recently been combined with spectroscopic studies leading to a successful separation of topographic, electronic, and magnetic information as well as magnetic domain and domain wall images of ferromagnetic rare earth and transition metal surfaces with subnanometer spatial resolution [7,8].

Here, we report on the first spin-polarized scanning tunneling spectroscopy (SP-STS) study of the surface of an antiferromagnetic bulk crystal, Cr(001), by making use of the spin-polarized d-like surface state located close to the Fermi level E_F. The surface state peak intensity in the local tunneling spectra is found to exhibit periodic alternations from one (001) terrace to the next, consistent with the topological antiferromagnetic order of the Cr(001) surface. For the first time, the effect of screw dislocations on the topological antiferromagnetic order could be studied with nanoscale spatial resolution. Such screw dislocations cause spin frustration in their surroundings leading to the formation of domain walls which are not observed in screw dislocation-free regions. The width of the domain walls is found to be on the order of 120 nm.

The experiments were performed with a commercially available ultrahigh vacuum (UHV) scanning tunneling microscope (STM) system operated at room temperature [9]. The Cr(001) single crystal has been prepared by repeated cycles of Ar^+-ion etching and annealing until a nitrogen- and oxygen-free surface was obtained as checked by Auger-electron spectroscopy (AES). A slight amount of carbon contamination was detected by AES which, however, did not affect the observation of the Cr(001) surface state peak by tunneling spectroscopy. Spin-resolved STM and tunneling spectroscopy studies were performed by using *in situ* prepared Fe-coated probe tips as described earlier [7]. Tunneling spectra were measured by adding a modulation voltage $U_{\mathrm{mod}} = 15\ \mathrm{mV_{rms}}$ to the applied sample bias U and recording the dI/dU signal by lock-in technique.

Figure 1(a) shows a constant-current STM image of a 500 nm $\times$ 500 nm surface region of Cr(001) including three different terraces separated by monatomic steps. The terraces are descending from the lower left part to the right part of the image. The electronic structure of this surface has been investigated by local tunneling spectroscopy. A peak in the $dI/dU(U)$ characteristics close to zero bias caused by the Cr(001) surface state in the minority spin band is usually observed if clean tip and sample surfaces have been prepared [10]. The slight difference regarding the surface state peak position found in this study compared to earlier experiments [10] might be caused by different amounts of residual impurities within the Cr(001) surface [11]. The intensity of the surface state peak is independent of the location on the Cr(001) surface if spin-averaged spectra are measured by using nonmagnetic probe tips such as tungsten. However, since the Cr(001) surface state is of minority-spin character [10], a spin-polarized

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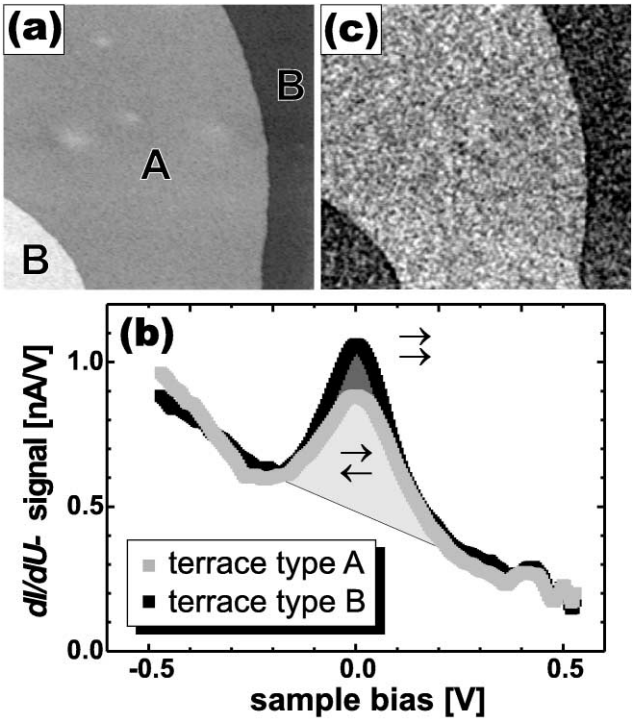

FIG. 1. (a) Constant-current STM image (500 nm × 500 nm) of the Cr (001) surface showing three different terraces separated by monatomic steps. The data were obtained with a Fe-coated probe tip using a tunneling current $I = 0.18$ nA and a sample bias $U = -60$ mV. (b) The two types of local tunneling spectra of Cr (001) obtained with a Fe-coated tip, spatially averaged over two different (001) terraces which are separated by a monatomic surface step. A clear difference in the intensity of the surface state peak close to E_F is found in contrast to spin-averaged local tunneling spectra obtained with nonmagnetic probe tips which always show the same surface state peak intensity. The two arrows symbolize the relative spin orientation of the tip (→) and the sample (← or →). (c) Spin-polarized spectroscopic image of the same surface region as shown in (a). The measured dI/dU signal at the surface state peak position changes between two levels whenever a monatomic step of the Cr(001) surface is crossed.

tunneling study with a ferromagnetic probe tip should be sensitive to the alternating magnetization of the (001) terraces. This is indeed observed experimentally. Figure 1(b) shows the two types of local tunneling spectra obtained in the constant-separation mode with a Fe-coated probe tip above two magnetically inequivalent (001) terraces. The stabilization parameters before the feedback loop of the STM was opened were $I = 0.3$ nA and $U = +700$ mV. A clear difference in the surface state peak intensity close to zero bias is measured for terraces with opposite spin orientation, i.e., types *A* and *B*. To prove the spatial correlation of the two different surface state peak intensities with the topography of the Cr(001) surface, we present a SP-STS image in Fig. 1(c) which has been obtained simultaneously with the topographic image of Fig. 1(a). In Fig. 1(c) the gray scale corresponds to the intensity of the spectroscopic dI/dU signal close to the surface state peak position ($U = -60$ mV) rather than to the topographic height as in Fig. 1(a). It is clearly seen that the spatial distribution of the spin-resolved spectroscopic signal is consistent with the model of alternately magnetized (001) terraces separated by monatomic steps. Because the data of Fig. 1(c) were measured in the closed-loop constant-current mode and at a bias voltage close to the surface state peak position ($U = -60$ mV), the tip approaches closer to the sample surface when located above a terrace of type *A* than above one of type *B*. As a result, the dI/dU signal becomes larger above terraces of type *A* than above those of type *B*, leading to a contrast inversion compared to spectroscopic dI/dU measurements in the constant-separation mode. This interpretation is supported by further data discussed below. A similar behavior has been observed for the Kondo resonance of Co atoms on Cu(111) [12] which is also located close to E_F.

In a second experiment we have looked at larger scale surface regions (1 μm × 1 μm) of Cr(001) as shown in Fig. 2(a). In this case, many different terraces of varying width, but always separated by monatomic steps are visible. In addition, three screw dislocations appear which are frequently observed at Cr(001) surfaces by STM [13]. Such screw dislocations should result in spin frustration in their surroundings and are therefore particularly interesting objects to study by SP-STS. Figure 2(b) shows the simultaneously measured spin-resolved spectroscopic image which again exhibits the dI/dU-signal intensity variations close to the surface state peak position over the different (001) terraces. In dislocation-free areas a spatially periodic dI/dU signal is obtained [Fig. 2(c), bottom part] with the periodicity given by the terrace width (about 80 nm in this case). Though the corresponding constant-current topography of Fig. 2(a) is primarily dominated by the terrace-and-step structure of the Cr(001) surface, the topological antiferromagnetic order is reflected by alternating measured monatomic step heights as demonstrated by the line profile of Fig. 2(c)(top part). This alternating apparent step height is a manifestation of the different tip-sample separations above terraces of types *A* and *B* which are a result of the spin sensitivity of the experiment. Since the conductivity dI/dU as measured in the constant-separation mode is higher (lower) above terraces of type *B* (*A*) due to an (anti)parallel relative spin-orientation between tip and sample the tip has to be withdrawn from (approached towards) the surface in order to keep the tunneling current constant. Consequently, the step height is reduced for an *A*-*B* step and enhanced for a *B*-*A* step. The correlation of the simultaneously measured topographic and spectroscopic signal as plotted in Fig. 2(c) nicely confirms our considerations regarding the differences between measurements performed in the constant-current and constant-separation mode mentioned above. More quantitatively, the measured step-height values differ periodically by about 0.14 Å. This variation is smaller by a factor of 3 than reported previously [5,6]

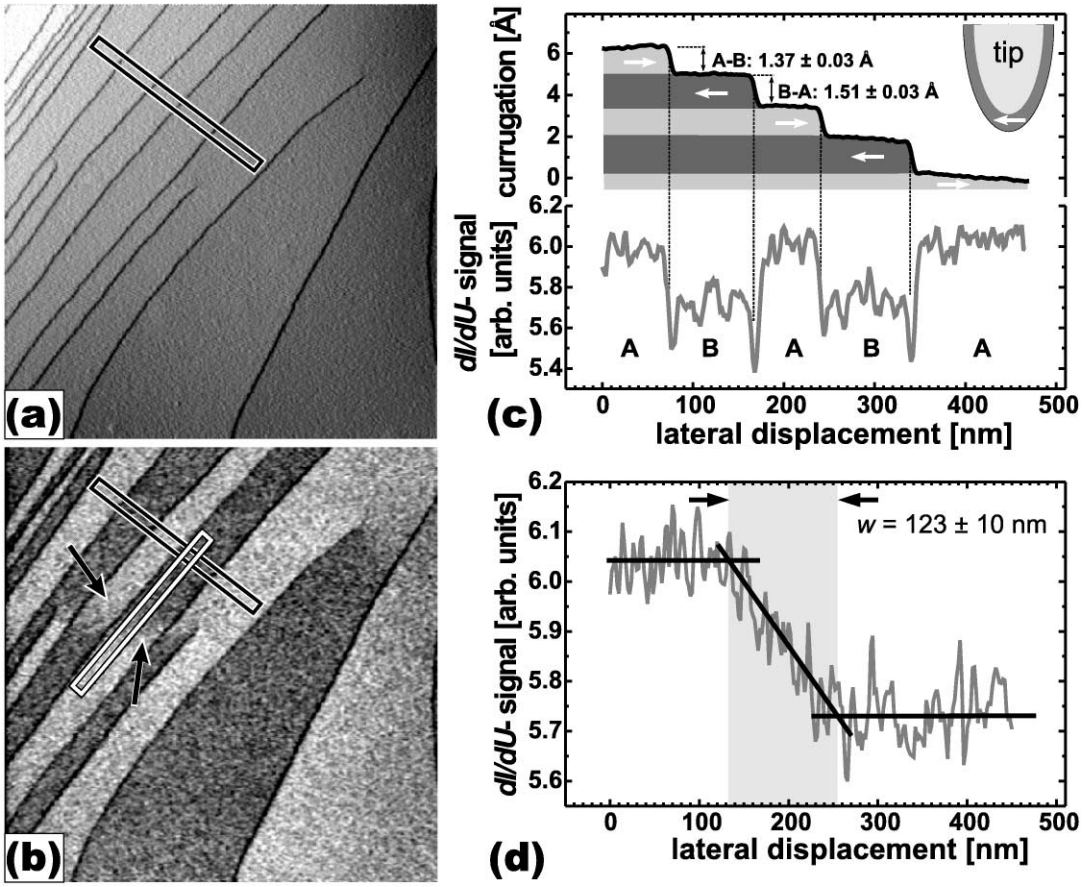

FIG. 2. (a) Large scale constant-current STM image (1 μm $\times$ 1 μm) of the Cr (001) surface showing many terraces separated by monatomic steps. Three screw dislocations are visible. The data were again obtained with a Fe-coated probe tip using a tunneling current $I = 0.18$ nA and a sample bias $U = -60$ mV. (b) Spin-polarized spectroscopic image of the same surface region as shown in (a). Again, the measured dI/dU signal at the surface state peak position changes between two levels whenever a monatomic step of the Cr(001) surface is crossed. However, an additional change can now be observed within (001) terraces in the surroundings of screw dislocations (see arrows). This is caused by spin frustration effects in the vicinity of such screw dislocations. (c) The top part shows a line section across five terraces separated by monatomic steps as measured with the Fe-coated tip in the constant current mode and statistically averaged over the surface region indicated by the box in (a). The step heights periodically alternate as found earlier by using CrO_2 tips [5]. The lower part shows the periodic alteration of the spectroscopic dI/dU signal, statistically averaged over the same black box indicated in (a) and (b). (d) Line profile across a domain wall statistically averaged over the white box shown in (b). The wall width is approximately 120 nm.

which, however, is expected since the degree of spin-polarization of the Fe tip is smaller than the one of CrO_2 tips as used in the earlier study [5,6]. By assuming a spin-polarization P_{Fe} of about 40% for the Fe-coated probe tip as deduced from earlier experiments [14] the measured step height difference of $2\Delta s = 0.14$ Å can be translated into a value for the spin polarization P_{Cr} of the Cr(001) surface. Making use of the relationship (4) from Ref. [5] we find $P_{\mathrm{Cr}} = 17\%$. This is consistent with the earlier result obtained by using CrO_2 thin film tips [5] if one assumes a polarization of the CrO_2 tip close to 100% which was indeed found by spin-resolved photoemission [15] and planar tunneling junction experiments.

We now focus on the spin frustration effects which occur in the surroundings of screw dislocations. As seen clearly in Fig. 2(b), the presence of screw dislocations leads to a change of the spin-resolved spectroscopic signal within a given (001) terrace which is never observed in dislocation-free surface regions. The spin frustration occurring in the surroundings of screw dislocations obviously results in the formation of domains and domain walls. This is most clearly seen on the two terraces indicated by arrows in Fig. 2(b). The line profile across such domain walls [Fig. 2(d)] reveals a wall width of approximately 120 nm. Since we have measured domain wall widths down to 2 nm in Fe nanowires [8] by using the same experimental technique we can conclude that the measured domain wall width of 120 nm on the Cr(001) surface represents an intrinsic wall width which is not broadened by instrumental effects. The possible formation of such domain walls in Cr(001) has been discussed previously in connection with the growth of Fe/Cr-bilayers (see, e.g., [3]). However, in this case the domain wall widths in the Cr layer are very much dependent on its thickness due to an

antiferromagnetic coupling of the Cr and Fe magnetic moments at the Fe/Cr interfaces [16]. Domain wall widths in bulk Cr have not been determined experimentally so far. Theoretical studies of domain walls in bulk Cr based on *ab initio* calculations predict wall widths on the order of 20 Cr lattice sites (i.e., about 3 nm) being reduced by a factor of 3 or 4 at the Cr(001) surface [17]. However, all calculations have been done assuming collinear spin orders, which is equivalent to an infinite anisotropy. This reduces the calculated wall width to the minimum. Therefore, it is not surprising that the experimentally determined wall widths are considerably larger. In fact, the value of approximately 120 nm as determined in the present study is of the same order of magnitude as the experimentally determined wall width of 210 nm at the Fe(001) surface by using scanning electron microscopy with polarization analysis (SEMPA) [18]. Since SEMPA has a probing depth which is much larger than the separation of the alternately magnetized (001) planes of Cr, this technique failed to observe the topological antiferromagnetic order of the Cr(001) surface. On the other hand, magnetic force microscopy (MFM) fails to reveal domains and domain walls in antiferromagnetic samples due to the lack of a measurable magnetic stray field of the sample which the magnetic probe tip can interact with. The present SP-STS study represents not only the first real-space observation of spin frustration effects and domain walls at the Cr(001) surface but also confirms that such domain walls already exist before Fe deposition on Cr(001) substrates [16].

In summary, the present study shows that spin-polarized electronic states at surfaces of antiferromagnets can be used for magnetic domain imaging similarly as for surfaces of ferromagnets [7,8]. Our SP-STS results are fully consistent with the presence of topological antiferromagnetic order at the Cr(001) surface [2,5]. In addition to previous theoretical and experimental studies, we have investigated for the first time the effect of spin frustration on the topological antiferromagnetism caused by the presence of screw dislocations. We have found that this spin frustration leads to the formation of domains and domain walls. The wall width has been determined to be on the order of 120 nm. We would like to point out that the spectroscopic dI/dU feature of Cr(001) close to zero bias can be observed down to the atomic level [19,20]. It is therefore possible to address fundamental questions of magnetism down to the atomic scale, e.g., during alloying [19] or during oxidation [20], by making use of spin-polarized tunneling spectroscopy. A recent SP-STM study of a two-dimensional antiferromagnet, a single monolayer of Mn on W(110), has proven that magnetic imaging of antiferromagnetic spin order is possible with atomic resolution [21].

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